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REMOTE SENSING APPLICATIONS OF PULSED PHOTOTHERMAL  
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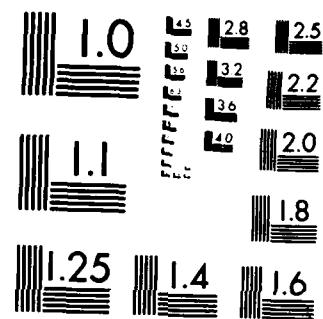
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A. C. Tam and B. Sullivan

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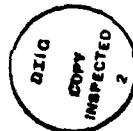
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**REMOTE SENSING APPLICATIONS OF PULSED PHOTOTHERMAL RADIOMETRY\***

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**ABSTRACT:** The technique of pulsed photothermal radiometry (*i.e.*, detection of the transient infrared thermal radiation from a condensed matter sample heated by a short-pulsed radiation) is useful for signal-ended remote sensing applications. We demonstrate for the first time such applications for spectroscopic detection, measurement of absolute absorption coefficients in opaque materials and sensing of dimensions or thermal properties in inhomogeneous materials. Theory of the PPTR technique in the simple case of a semi-infinite homogeneous material is described.

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## REMOTE SENSING APPLICATIONS OF PULSED PHOTOTHERMAL RADIOMETRY\*

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Optical absorption coefficients are typically measured by conventional transmission measurements (TM). However, TM is reliable only in the medium absorption range, *e.g.*, with absorption coefficients  $\alpha$  in the range of  $\sim 10^{-2}$  to  $\sim 10^2 \text{ cm}^{-1}$ . For very weak absorptions (*i.e.*,  $\alpha \ll 10^{-2} \text{ cm}^{-1}$ ), TM is frequently unreliable.<sup>1</sup> On the other hand, for very strong absorptions ( $\alpha \gg 10^2 \text{ cm}^{-1}$ ), little or no light can penetrate through a sample of typical thickness. Furthermore, TM cannot be used for remote sensing since it is a double-ended<sup>2</sup> method. We describe in this paper the new method of pulsed photo-thermal radiometry (PPTR) that has the following advantages: (1) Absolute optical absorption coefficients may be obtained for very opaque samples (*i.e.*,  $\alpha > 10^3 \text{ cm}^{-1}$ ); (2) PPTR is a single-ended<sup>2</sup> measurement technique utilizing intense pulsed laser for excitation; thus, it can be used as a new mode of LIDAR<sup>2</sup> (Light Detection and Ranging) for remote sensing applications.

The principle of PPTR is the following. An excitation laser pulse of short duration is absorbed by the sample, causing photo-thermal heating, *i.e.*, optical absorption with subsequent heating by thermal de-excitation. This causes an increased "black-body radiation" from the sample, and a suitable infrared (IR) detector is used to record this transient IR signal. To achieve a single-ended detection scheme, both the excitation laser and the IR detector are on the same side of the sample. Our PPTR technique is different from the flash thermal diffusivity measurement of Parker *et al.*<sup>3</sup> and Deem and Wood<sup>4</sup> because these earlier measurements are double-ended, and also no absolute absorption coefficient were obtained. It also differs from the recent developments of continuous

modulated photothermal radiometry (CMPTR) techniques.<sup>5,6,7</sup> In CMPTR, a continuous laser beam modulated at about 50% duty cycle is used for excitation, and the magnitude and phase of the IR radiometry signal can be measured; the information is usually insufficient to obtain absolute absorption coefficients. Furthermore, the relatively large duty-cycle produces substantial unmodulated heating effect, which may affect signal interpretation. With the use of intense pulsed laser sources, PPTR (rather than CMPTR) should be useful for long-distance remote sensing. However, limitations on the usefulness of PPTR occurs for samples of low IR emissivity or low laser-damage thresholds.

The mathematical basis for the present PPTR technique has been given by Carslaw and Jaeger.<sup>9</sup> Consider a semi-infinite solid or liquid material with its free surface being defined by  $x=0$  (the positive  $x$ -axis goes normally into the material). Suppose at time  $t=0$ , the (initial) temperature profile of the material is described by a function  $f(x)$ . The subsequent temperature profile  $\theta(x',t)$  is (Carslaw and Jaeger,<sup>9</sup> p. 53)

$$\theta(x',t) = (\pi Dt)^{-1/2} \int_0^{\infty} f(x) \exp[-(x-x')^2/4Dt] dx \quad (1)$$

where  $D$  is thermal diffusivity of the material. In our experiment, the initial temperature distribution  $f(x)$  is produced by a short-pulsed optical absorption at  $t=0$ :

$$f(x) = A \exp(-\alpha x) \quad (2)$$

where  $A$  is the initial surface temperature and  $\alpha$  is the optical absorption coefficient. The temperature increase  $\theta(x',t)$  produces<sup>5-8</sup> a PPTR signal  $S(t)$  given by

$$S(t) = K \alpha' \int_0^{\infty} e^{-\alpha' x'} \theta(x',t) dx' \quad (3)$$

where  $K = 4\epsilon\sigma T^3$  is a constant,<sup>5</sup>  $\epsilon$ =emissivity,  $\sigma$ =Stefan-Boltzmann constant,  $T$ =absolute temperature and  $\alpha'$ =the IR absorption coefficient of the sample averaged over the detection IR bandwidth. Combining Eqs. (1)-(3), we get

$$S(t) = AK\alpha'(\pi Dt)^{-1/2} \int_0^\infty \int_0^\infty \exp[-\alpha x - \alpha' x' - (x-x')^2/4Dt] dx dx'. \quad (4)$$

While Eq. (4) can be used in general to relate the observed PPTR signal  $S(t)$  with the optical and IR absorption coefficients and the thermal diffusivity, we can best illustrate its usefulness for the simplified case when one of the absorption coefficient is much larger than the other, e.g.,  $\alpha' \gg \alpha$ . In this case,

$$\begin{aligned} S(t) &\approx AK(\pi Dt)^{-1/2} \int_0^\infty \exp[-\alpha x - (x^2/4Dt)] dx \\ &= AK \exp(t/4\tau_0) (1 - \text{erf} \sqrt{t/4\tau_0}) \end{aligned} \quad (5)$$

where  $\tau_0$  is the thermal diffusion time for a distance  $\alpha^{-1}$ :

$$\tau_0 = \alpha^{-2}/4D \quad (6)$$

and "erf" is the "error function".<sup>9</sup> Equations (5) and (6) indicate that the optical absorption coefficient  $\alpha$  can be obtained from  $S(t)$  if  $\alpha' \gg \alpha$ . In the general case where  $\alpha' \gg \alpha$  is not valid, the PPTR signal can also provide a measure of  $\alpha$  or  $\alpha'$  by using the general Eq. (4).

Our experimental arrangement to demonstrate the technique of PPTR for quantitative remote sensing applications is shown in Fig. 1. The excitation laser pulse is obtained from a flashlamp pumped dye laser (Candela LFDL-1). The laser wavelength is about 590 nm (with about 10 nm bandwidth), energy is about 5 mJ, pulse duration is about 1.5  $\mu$ sec

(full-width at half-maximum), and repetition rate is about 2 Hz. The excitation laser spot at the sample surface has a diameter of 0.5 cm. The P PTR signal is collected by a gold-coated spherical reflector, and detected by an infrared detector fitted with a Ge window which transmits in the 3-11  $\mu\text{m}$  range. The liquid-N<sub>2</sub> cooled detector (New England Research, MPC11-2-B1) made of a 2mm  $\times$  2mm HgCdTe crystal sensitive to 8-12  $\mu\text{m}$  radiation, has a rise time of about 200 nsec. The detected P PTR signal is amplified by a broad band preamp (Ithaco model 1201) and stored in a signal-averaging oscilloscope (Tektronix 7854). Highlights of some of the samples studied, and some of the first results are given below.

**1. Homogeneous Opaque Solid Sample.** The sample is a piece of opaque black rubber of thickness about 0.5 cm. The observed P PTR signal is indicated in Fig. 2(a). The fitting of the signal to Eq. (5) is shown in Fig. 2(b) with  $\tau_0$  being 5  $\mu\text{sec}$ , which implies (through Eq. (6)) that the absorption length  $\alpha^{-1}$  of the black rubber sample at 590 nm is 1.4  $\mu\text{m}$  (corresponding to an initial surface heating of  $\sim 70^\circ\text{C}$ ). To verify the  $\alpha^{-1}$  value, we use a "Microtome" sectioning system (LKB Ultrotome III) to make a 3  $\mu\text{m}$  slice of the sample; the 590 nm transmission through this rubber slice on a quartz substrate is about 10%, indicating an absorption length  $\alpha^{-1}$  of 1.3  $\mu\text{m}$ . This experiment clearly shows for the first time that the P PTR technique is useful for the remote measurement of absolute absorption coefficients.

**2. Homogeneous Opaque Liquid Sample.** The observed P PTR signal for a highly opaque liquid (Patent Blue A dye dissolved in water at a concentration of 10% weight/volume) is shown in Fig. 3(a). The theoretical fit by Eq. (5) (see Fig. 3(b)) yields an absorption length  $\alpha^{-1}$  of 7  $\mu\text{m}$  (corresponding to an initial surface heating of  $\sim 20^\circ\text{C}$ ). The direct transmission measurement of  $\alpha^{-1}$  is difficult; we do this by putting the liquid in a 100  $\mu\text{m}$  path-length glass cell, and measure the transmission of the laser beam. We obtained  $\alpha^{-1}$  to be approximately 3  $\mu\text{m}$  with an uncertainty of factor of 3.

**3. Layered Materials.** So far, we have only considered homogeneous materials. However, PPTR can also be used for remote measurement of layered materials, which may produce characteristic features on the time-development of the PPTR signal. To show this, we put a thin transparent polyester coating (thickness about  $45 \mu\text{m}$ ) onto the black rubber sample studied above. The observed PPTR signal is shown in Fig. 4. For the coated sample, the PPTR signal shows an initial fast decay (due to the IR emitted from the black rubber surface, and transmitted through the coating), and subsequently a peaked signal at late times (due to the heat diffusing from the rubber surface). This permits us to derive the coating thickness  $\ell$  from the thermal diffusivity  $D$  of the coating ( $D \approx 1 \times 10^{-3} \text{ cm}^2/\text{sec}$ ) and the time  $t_{\text{pk}}$  of the diffusion peak of the PPTR signal ( $t_{\text{pk}} \approx 8 \times 10^{-3} \text{ sec}$ ), using a one-dimensional thermal diffusion formula:<sup>10</sup>

$$\ell = [2Dt_{\text{pk}}]^{1/2} = 40 \mu\text{m} . \quad (1)$$

This compares very well with a direct measurement of  $45 \mu\text{m}$ , thus, showing the capability of single-ended remote measurement of thickness or thermal properties of layers by PPTR.

**4. Agglomeration of Powders.** Thermal conduction from one powder particle to another depends on the state of agglomeration of the powder sample. To show how this affects the PPTR signal, we study two black carbon-loaded epoxy powder samples, one being loosely-piled and unconsolidated, and the other being compressed and consolidated. The observed PPTR signals shown in Figs. 5(a) and (b) respectively clearly suggest the strongly hindered heat transport from one particle to another in the former case.

In conclusion, we have made the first quantitative experimental study of single-ended pulsed photo-thermal radiometry techniques for remote spectroscopic detection, measurement of absolute absorption coefficients and sensing of physical or thermal properties of samples.

Applications presently demonstrated include absolute absorption spectroscopy in solids and liquids, sensing of layered materials and monitoring of state of agglomeration of powders. Other applications include remote identification of aerosol samples and remote sensing of chemical reactions, polymerizations, sedimentation, *etc.*, and will be reported in future publications.

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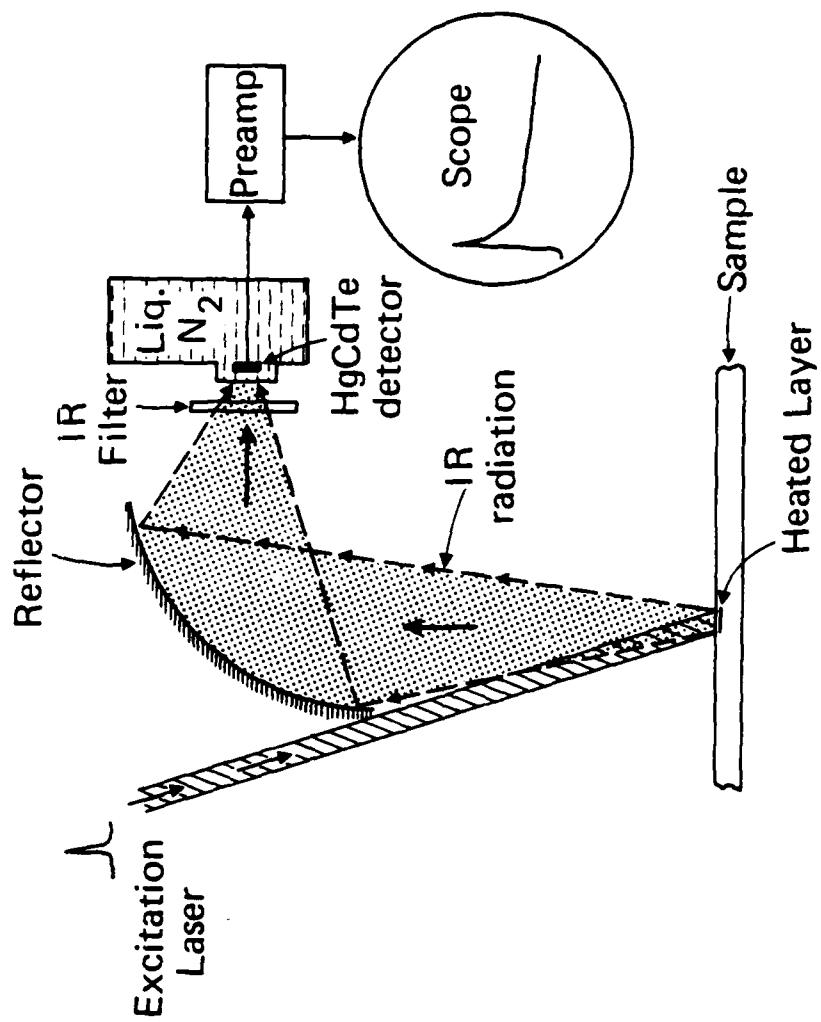


Figure 1. Schematics of the experimental apparatus for pulsed photo-thermal radiometry (PPTR) that is useful for remote sensing applications.

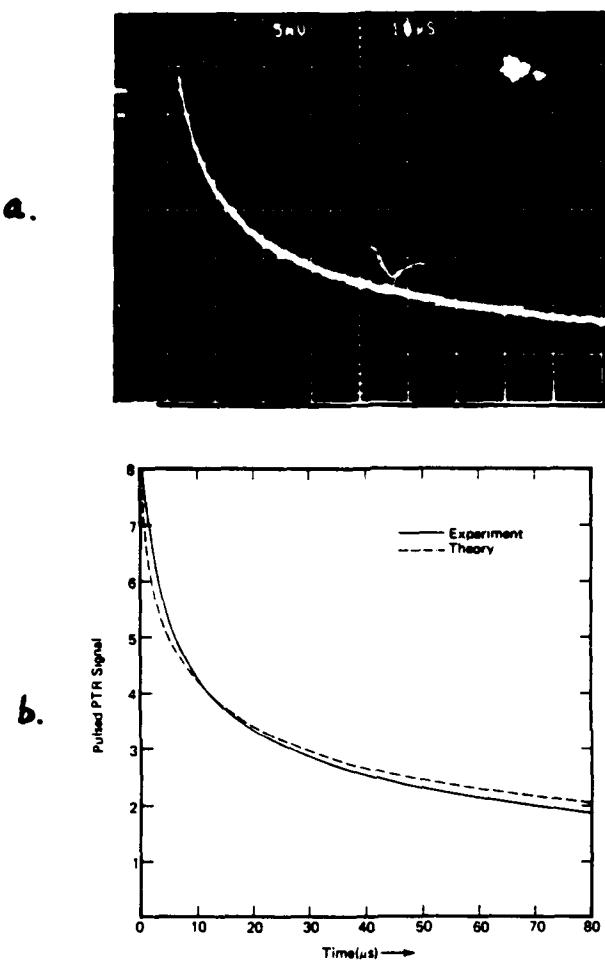
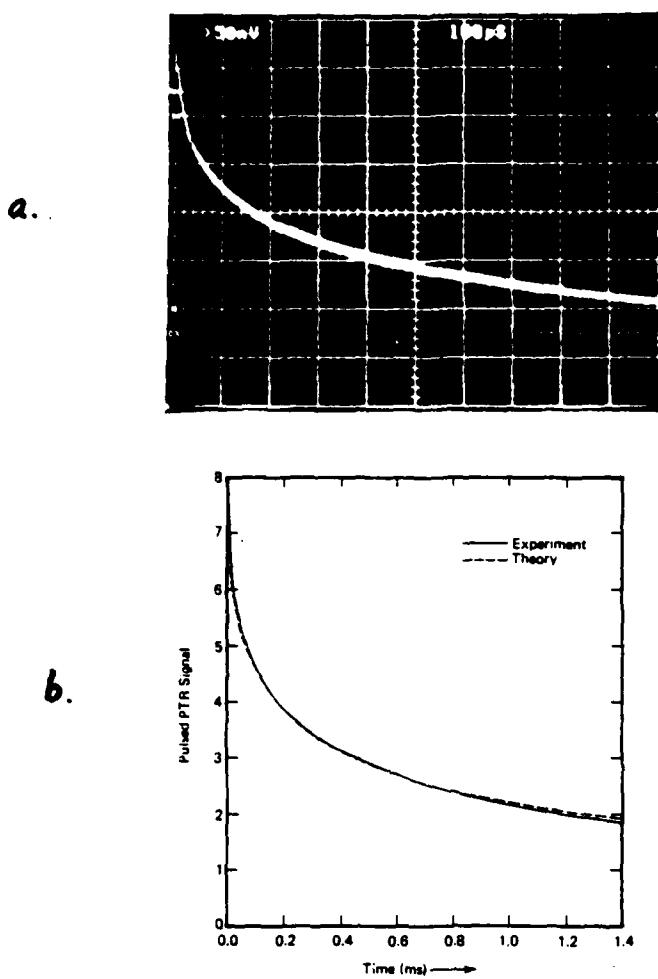


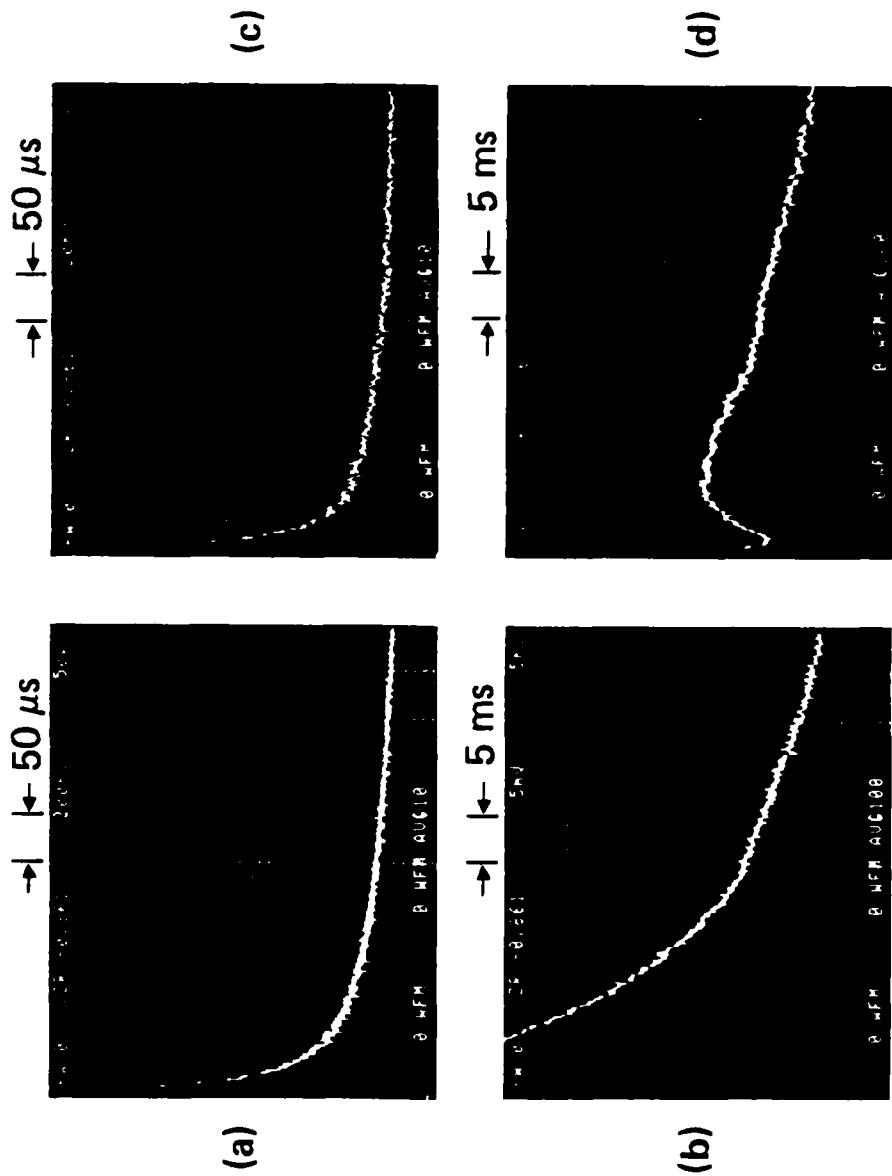
Figure 2. (a) Observed PPTR signal for a black rubber sample. Horizontal scale: 10  $\mu$ s/division.

(b) Fitting of observed signal to Eq. (5).

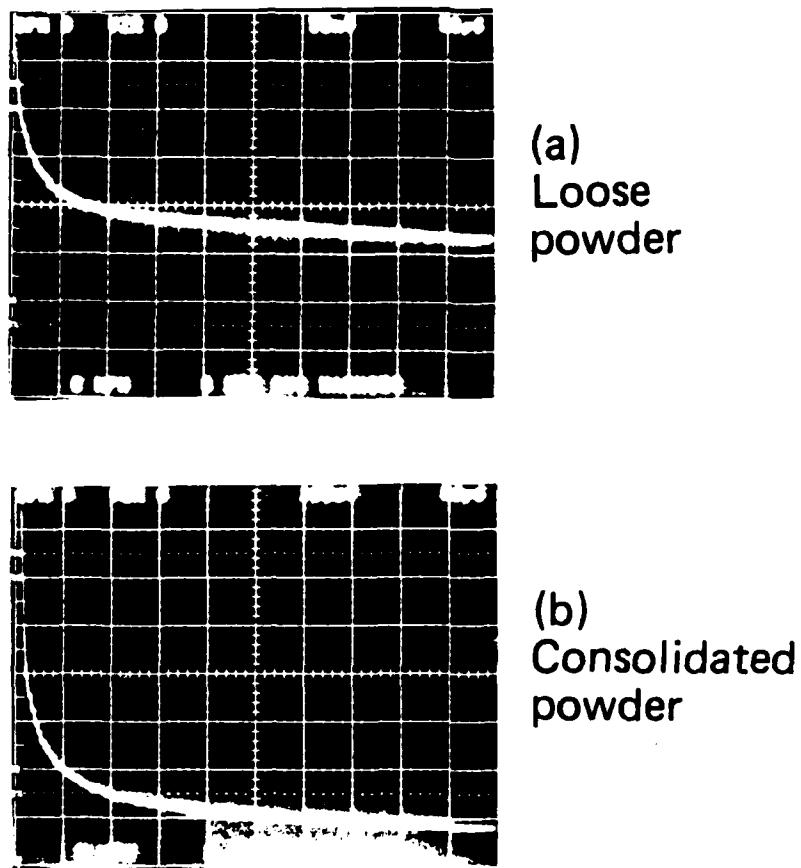


**Figure 3.** (a) Observed PPTR signal for a strong dye solution. Horizontal scale: 100  $\mu$ s/division.

(b) Fitting of observed signal to Eq. (5).



**Figure 4.** Observed PPTTR signal for a bare black rubber sample (a and b) compared to the same sample coated with 45  $\mu\text{m}$  of optically transparent polyester (c and d). Verticle sensitivity in (b) is 40x that in (a), and in (d) is 4x that in (c).



**Figure 5.** Observed PPTR signal for (a) unconsolidated and (b) consolidated carbon-loaded epoxy powder. Horizontal scale: 50  $\mu$ s/division. The bottom of each scope picture is the zero signal level.

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